1. INTRODUCTION

Color is one of the distinct elements of nature, which has made human life more beautiful, attractive and charming throughout the ages. In ancient civilization, people tried to decorate themselves with natural colors around them with inspiration from plants, flowers and various other sources. Due to the urge for beauty these colors were born from natural origin. The colour derived from different natural resources, such as flowers, vegetables, fruit residues, plants, animals and minerals, are called natural pigments[1]. Natural colors were the only available colours up to 1850[2]. Later, with the invention of synthetic colors, it took very little time for the natural dye to be obsolete from the market. The whole market was captured by synthetic colors, because of their abundant availability, low cost, application simplicity and better colouring properties than natural colors. There are wide range of colors and shades available in synthetic domain. Synthetic pigments dominated the market by almost half a century. However, at the end of the 20th century, many countries of Europe and Asia including India banned certain synthetic pigments due to their harmful impact on nature and their habitants[3]. In the 21st century, an increasing awareness towards environment has pushed scientists and researchers to start work on natural dye sources. Recently, the use of natural dye is increasing due to various stringent environmental standards imposed by various European and Asian countries including India[4].

Natural colours have some drawbacks such as poor reproducibility, limited color, non uniformity of colors and inadequate color stability and two-phase application (Mordanting and dyeing) process[5]. Still it has some very special advantages which are environmental-friendliness, renewability of their sources, minimal consumption of synthetic chemicals and unusual but soothing color shades[6]. Also these natural colorants do not cause health hazards and produce a rich variety of colors[7].

Some natural pigments are fugitive and require a mordant to increase their dye uptake and colour fastness. Mordants form more functional groups on the cloth than untreated one. Those groups provide more reactive sites to engage dye molecules, and therefore improve the dyeing efficiency as well as their fastness properties. Some metallic salts such as alum ($KAl(SO_4)_2$), stannous

chloride ($Cl_2H_4O_2Sn$), stannic chloride ($SnCl_4$), ferrous sulphate ($FeSO_4$), copper sulphate ($CuSO_4$) and potassium dichromate ($K_2Cr_2O_7$) are widely used as mordants[8]. Dyer uses different types of natural colors, based on their fixation properties, for the dyeing of various textile materials in different forms. Various textile materials includes wool, cotton, silk etc and in different forms like fibre, yarn, fabrics and other textile products.

An attempt has been made to explore the newly popularized research field of traditional natural dye in the following literature review. In this segment various prospects of natural dye is being investigated or scrutinized as per the available modern research literature.

2. LITREATURE REVIEW

2.1 Colour and Colouring Materials

Electromagnetic radiation is a form of energy. It can be classified into different types as per the wavelength. Visible light ranges from 400nm to 700nm (Fig 1) in the full spectrum of electromagnetic radiation range. Colour is the phenomenon of any visual photo chemical experience of a particular wavelength. In nature white light defines presence of all colours. They are additive in colour. White light is the combination of three primary colours i.e. red, blue and green. When the equal part of these three primary colours mixed then white colour is formed[10].

While in macro material world colour evolves with three specific interactions between visible range electromagnetic waves and material. Those three interactions are reflection, refraction and absorption. During these interactions a specific range or wavelength of colour get reflected or transmitted. That very phenomenon decides the colour of any material. Therefore colour of any macro material without any self emission ability in the visible range acquires colour which are subtractive in nature. There are three primary subtractive colours i.e. cyan, magenta and yellow. When the equal part of these three primary subtractive colours mixed then black colour is formed.

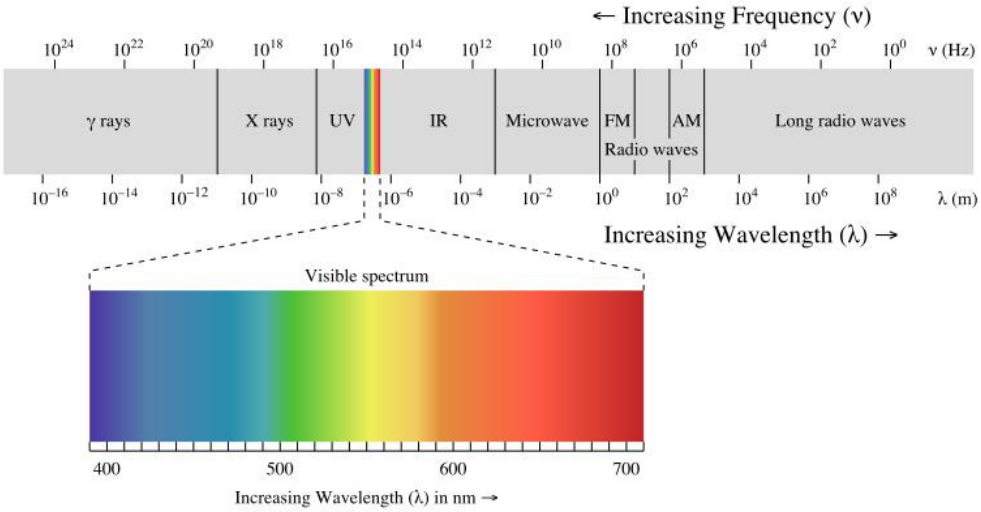

Figure1. Electromagnetic spectrum[9].

In chemical world unsaturated bonds usually provides colour. They can be categorized mainly in i.e. dyes, pigment, and lakes (ingrain color). Each category is briefly discussed below.

2.1.1 Dyes

Dye is as substance which is used to impart colour on various material like paper, leather, textile material like fibre, yarn of fabrics and which cannot be easily removed from them. The dye structure mainly consists of three parts - chromospheres, chromogen and auxochrome[10].

- Chromospheres: The chromospheres is an unsaturated group that absorbs light and reflects the same at particular angle to provide hue, eg, keto, azo, nitroso, nitro, thio, ethylene etc.
- Chromogen: Chromogen retains chromospheres which play a key role in determining the final hue and its affinity towards fiber, its stability, fastness properties etc.

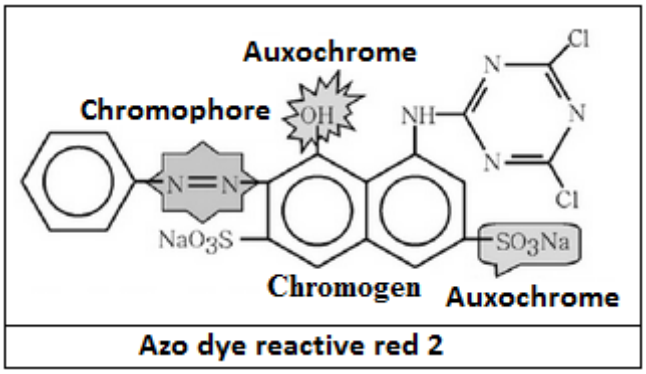

Figure 2. Azo dye reactive red 2[11].

- Auxochrome: Auxochrome are acidic or basic groups which are there in the dye structure and are responsible for deepen the shade, e.g. –OH (Hydroxyl group), SO_3H (sulphonic group), - COOH (carboxylic group), - NH_2 (amino group) etc.

  Example of azo dye reactive red 2 showing chromophore and auxochrome are given above in Figure 2.

Dye can be classified in terms of source and chemical structure. As per the sources dyes are natural and synthetic. While as per chemical nature dyes are classified as direct dye, acid dye, basic dye, azo dye, vat dye, reactive dye etc. Natural dyes are obtained from the plant, animal or mineral origin. The synthetic dyes are manufactured from chemicals. Direct dyes can directly be applied on the fabric in a hot dye bath with or without mordants. They are generally applied to cellulosic fibre material. They have poor washing and light fastness. Acid dyes and basic dyes are used to apply on natural protein fibres and also synthetic material like nylon, acrylic etc. They have poor washing fastness but their light fastness is fair to good. Azo dyes can be applied to both cellulosic and protein fibres and have excellent washing and light fastness. Vat dyes are applied on cellulosic fabrics by two step process. First dye is reduced to make it solublised and then after application of it on the fabric it oxidized in the air to get excellent fastness properties. Reactive dyes are directly react with the fibre to form bond and gives excellent fastness rating. It can be applied to wide range of fabric which includes cotton, wool, silk etc[10].

2.1.2 Pigments

Pigment is an insoluble finer powder that makes the color by its reflection of specific spectral wavelength. It has an excellent coloring ability. It is applied to the surface of the material only[10]. Difference between dyes and pigments are given in the following table. (Table 1.)

Table 1. Difference between dyes and pigments.

S. No.	Dyes	Pigment
01	Dyes are normally water solubility.	Pigments are insoluble in water.
02	Most of all the dyes are organic compounds and the remaining are metallic compound.	Most of the pigments are inorganic compound.
03	Dye diffusion in the fabric.	Pigment diffusion on the fabric.
04	It's not possible to dye all types of fabric by using dye.	All types of fabric can be dyed by using pigment.
05	No binding agent is required in case of dye.	Binding agent is required in case of pigment.
06	It is costly as these prepared chemically.	It is cheap as these found by mineral origin.

2.1.3 Lakes (Ingrain colours)

Lake colours are normally insoluble in water and it can applied as a colour through dispersion. It is available mostly in powder form and can give very bright and consistent colour even if used in very small quantity. Lake colours are prepared by dispersing it in oils. They are ideally used as food colouring agents. Lake Colour is available in natural as well as synthetic colour. Basically two types of lake colours are commonly available. They are organic lake colours and aluminium lake colours [10].

The main objective of the proposed research revolves around natural dyes. Therefore, a critical review has been done in the following segment.

2.2 Natural dyes

Natural dyes are the natural colour obtained for the sources available in nature. The natural sources include plants, animals and minerals. Some examples of natural dyes are indigo dyes, henna dye, madder dye etc.

2.2.1 Natural Dye Sources

The natural colour can be obtained from various natural sources available in the nature. The natural dye source includes plants, animals, minerals and microbial sources. However plant is the major source of natural dyes which includes vegetables, vegetable skins, fruits, fruit skins, flowers, plant leftover, leaves, roots, husk, rind, bark etc[12]. A brief account of natural dye sources is consolidated in table 2 (a) and 2 (b).

Table 2(a): Natural dye source[13].

Botanical Name	**Common Name**	**Colour**
1. Bark Dye		
Acacia farnesiana Wild.	classic flower, Grand Babul	Black
Acacia leucophloea Wild.	Safed Kikar	red
Acacia sinuate (Lour) Merril	Ritha	Brown
Albizia odoratissima	Kala Siris	Brown
Careya Arborea Roxb.	Kumbhi	Brown
Emblica officinalis Gaertn.	Amla	Black, Grey and Brown
Lannea coromandelica (Houtt.)	Jingan	Golden and Pale brown
Mangifera indiac Linn.	Mango, Aam	Yellow
Morinda tinctoria Roxb.	Ach	Red
Pinus roxburghii Sarg.	Long-leaved pine chir	Yellow
Pinus wallichiana A.B. Jackson Orange	Indian Pine, Kail	Yellow to deep
Rhizophora mucronata Lam	Mangrove	chocolate
Shorea robusta Gaertn.f.	Sal Red and Black	Golden and Pale brown
2. Flower Dye		
Butea monosperma Lam.	Taub. Dhak, Palas	Yellow
Cassia auriculata Linn.	Tarwar	Yellow

Table 2(b): Natural dye source[13].

Botanical Name	Common Name	Colour
Crocus sativus Linn	Saffron, Kesar	Yellow
Gossypium herbaceum Linn	Cotton, Kapas	Yellow
Memecylon umbellatum Burm.	Iron wood tree	Yellow
Nyctanthes arbor-tristis Linn.	Harsinghar	Yellow
3. Fruit Dye		
Aegle marmelos (Linn.) Correax Roxb.	Bel	Yellow
Annona reticulata Linn.	Ramphal	Red
Diospyros Peregrine (Gaertn.)	Gab Gurke	Brown
Terminalia bellirica Roxb.	Bahera	Yellow
4. Leaf Dye		
Acacia nilotica willd	Kikar	Black to Dark
Anogeissus latifolia Wall	Bakla	Black
Cedrela toona Roxb.	Tun	Black
Garuga pinnata Roxb.	Chogar	Red
Lawsonia inermis Linn	Mehendi	Red
Tectona grandis Linn.	Asoka	Brown
Sarcochlamys pulcherrima	Teak, Sagwan	Yellow
Ziziphus mauritiana Lam.	Indian Jujube	Red- Pink
5. Root Dye		
Aleurites moluccana willd	Jangli Akhrot	Brown
Artocarpus lakoocha Roxb.	Dahua	Yellow
Berberis nepalensis Spreng	Nepal Barbeery	Yellow
Curcuma longa Linn.	Haldi	Yellow
Morinda Citrifolia Linn.	Indian mulberry	Red
6. Seed Dye		
Bixa orellana Linn.	Annatto	Orange and Red
Cassia Tora Linn.	Chakunda	Blue or Red
7. Wood Dye		
Acacia catechu Willd.	Khair	Red and Brown
Artocarpus heterophyllus Lam.	Kathal	Yellow
Caesalpinia sappan Linn.	Sapan wood, Bakam	Red
Moringa oleifera Lam	Soanjna	Blue
Pterocarpus santalinus Linn.	Sandalwood, Lal chandan	Red

2.2.2 Extraction Process of Natural Dye

2.2.2.1 General Method of Natural Dye Extraction

The dye is extracted from the natural dye source either from freshly chopped source or dried powdered form. The extraction is done in either aqueous condition by using distilled water or normal water or by using organic solvents like alcohol[15]. Through aqueous extraction method the dye is obtained from solution or in the powdered form. After fractionation of those precursors pure dye is obtained.

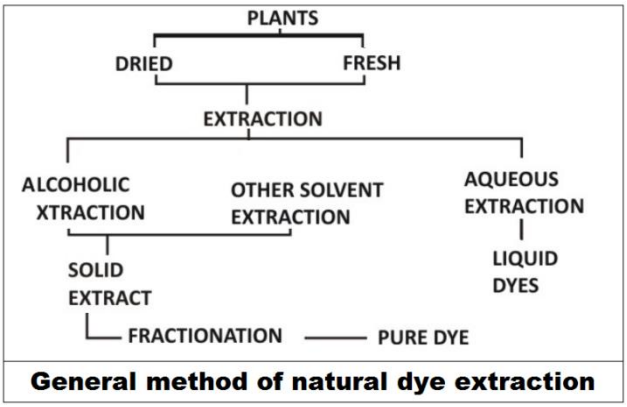

Figure 3. Method of natural dye extraction from plants[14].

The efficiency of the colour extraction from the natural colour source like plant, animal and mineral source depends on the type of extraction media used to extract dye (i.e. aqueous, organic media, acid or alkali). Also it depends upon the extraction conditions such as extraction time, temperature, pH of extraction bath and material to liquor ratio[16]. In the following segments aforementioned influencers are discussed in brief.

2.2.2.1.1 Effect of Extraction Media on Natural Dye Extraction.

2.2.2.1.1.1 Aqueous Extraction

In aqueous extraction methods distilled or normal water is used to extract the dye. The temperature range varies from 60^{o}C to 90^{o}C and sometimes boiling temperature of the extraction medium is used. Time of heating is kept between 60 min to 2 hrs and material

to liquor ratio is normally kept between 1:10 to 1:20[17]. Many scientists have done work on extraction of natural dyes through aqueous extraction. Janani *et al.*[18] studied the aqueous extraction medium based dye from coffee and mulberry leaves. They applied on silk fabric by using different mordants. They selected various mordants such as wood ash water, iron rust (Fe_2O_3), water, lemon juice and cowdung for dyeing the silk fabric. Good colour fastness was observed. Das *et al.*[19]extracted the dye from badam fruit peel and grandiflora flower by aqueous extraction method at 100^{o}C and applied on silk and cotton fabrics. The fabrics were dyed using copper sulphate as mordant and found good colour fastness properties (between 4-5 ratings). Pan *et al.*[20] extracted the dye from the leave of deodara, jackfruit and eucalyptus in soft water. Small pieces of the leaves were used and heated at boiling temperature for 4 h.

2.2.2.1.1.2 Organic Media Extraction

As organic extraction medium, various organic solvents such as ethanol, methanol, petroleum ether, acetone, chloroform and mixture of these with or without the presence of water are used for natural dye. The type of organic extraction media depends on the type of natural source. The extraction method which uses the mixture of water and alcohol is able to extract both water-soluble and water-insoluble substances from the natural dye sources. The extraction yield in organic media is higher than the yield of the aqueous method due to larger quantity of coloring materials can be extracted in former method. Many scientists have done work on extraction of natural dyes through organic media. Divya *et al.*[21] extracted the dye in ethanol from the fine powder of *Bixa orellana* seeds and sappan bark in soxhlet apparatus at 78^{o}C for 3 days. Srivastava *et al.*[22] extracted the dye colour from aloe vera leaf using 3:2 ethanol and water solution. Extraction was done in orbital shaker (at room temperature) for 24 hours. Further the dye was extracted by evaporating the solvent through evaporation by keeping the system open for 24 h. Thiyagarajan *et al.*[23] have extracted the dye by 4:1 solution of ethanol and water in soxhlet apparatus at 70^{o}C for 60 min. Kumar *et al.*[24] extracted the dye from waste of

food industry (i.e. from lemon and orange peel) in ethanol. They have found a higher dye extraction concentration in comparison to aqueous counterpart.

2.2.2.1.2 Effect of Extraction Bath Conditions on Dye Extractions

Extraction bath condition includes extraction time, extraction temperature, pH of extraction bath and material to liquor ratio of dye extraction bath. As the extraction time increases the percentage of extracted natural dye also increases upto a certain point of time and then it become saturated. The extraction tile ranges normally from 60 min to 90 min. Most utilized extraction temperature range is from 40^{o}C to 90^{o}C[16]. Sometimes extraction is done even in boiling temperature of the used medium. Effect of heat upon dye extraction is very prominent. It shows improvement till the point where depletion of dye overpowers the dye yield. Effect of pH depends upon the natural dye source and extraction condition. A huge amount of literature is available on the effect of extraction condition on dye extraction from different dye sources[17]. Sabarudin *et al.*[25] had studied the effect of extraction time and material to liquor ratio on dye extraction from bougainvillea flower *bract* and found the 60 min as optimum extraction time and 1:10 MLR is optimized. Arwati *et al.*[26] investigated the effect of extraction time on dye extraction from the jackfruit wood powder and found optimum dye extraction time as 2 h. Rangi *et al.*[27] studied the effect of extraction temperature and MLR ratio on dye extraction from Arjuna bark and found optimum extraction temperature as 100^{o}C and 1:20 MLR as optimum.

2.3 Identification of Natural Dyes

There are various methods used for the identification of natural dyes which includes thin layer chromatography (TLC), high performance liquid chromatography (HPLC), infra-red spectral studies and UV/visible spectroscopic studies. Through these methods one can easily identify a colour potential of particular natural source along with the chemical identification of the same[28].

Thin layer chromatography (TLC) technique is used to separate the compound present in natural dyes. For the separation of compounds two phases i.e. stationary phase and mobile phase are used. Silica gel is used as stationary phase and different solvent like ethanol, acetone and water are used as the mobile phase[29].

High performance liquid chromatography (HPLC) is the advanced technique used to separate and identify the compounds present in the natural dyes. This process also involves two phases for the separation. The stationary phase is a column of packing material (inert silica gel) and mobile phase is a gas. Dyes which cannot be analyzed by TLC can be easily analyzed by HPLC[30].

Infra-red spectroscopic study is the technique which is used to identify the functional group present in the organic compound (natural dyes). It is used to identify the samples in solid, liquid or gas form. In this process different mode of vibration is created in the compound with the help of infra red rays. Then the samples are identified through stretching and bending responsible for a particular compound[30].

UV/visible spectroscopic study are used for the qualitative analysis of the compounds present in the natural dyes. The aforesaid method involves excitation of the compound up to the exited level to obtain the absorption spectra. This is followed by the return of the compound to its ground state. During the return phase it emits the energy in the form of spectral lines which is called as emission spectra. This spectrum helps in finding the compounds present in the natural dyes. UV/visible spectroscopy are based on the principle known as Beer-Lambert law. Beer Lambert law is used to calculate the concentration of the dye solution using the given formula[31]

$$\text{Log } (I/I_0) = \epsilon dc \quad \text{----------} \qquad (1)$$

Where,

I = Intensity of the light source

I_0 = Intensity of the light passed through the solution

ϵ = Molar absorptivity or molar absorption coefficient.

d = Length of solution the light passes through

c = Concentration of the dye solution

2.4 Shelf Life of Natural Dye

Shelf life is considered as the length of time that a commodity can be stored without becoming unsuitable for use, consumption and sale[32]. It can be applied to foods, beverages, cosmetics, natural dyes and any perishable item. In other words, recommended maximum time for shelf life for which the products can be stored, during which the defined quality of the goods is acceptable under the required (or specified) conditions of distribution, storage and display[33]. Shelf life is influenced by so many factors like exposure to contamination by various agents and parameters such as heat, light, moisture and micro organisms[34]. Natural dyes have also their shelf life. Natural dyes which are remained unused for a long period of time is often get contaminated by microbes. Those infected dyes are unfit for use. Preservatives and antioxidants may be incorporated into natural dyes to extend their shelf life. Some work has been reported on shelf life of natural colour that is mostly used for food items. Marpudi *et al.*[35] reported the work on shelf life improvement of papaya fruit using antimicrobial coating based on aloe vera as preservative. Yadav *et al.*[36] have worked on shelf life improvement of mulberry juice by using various preservatives combination in various ratio. Verma *et al.*[37] studied the effect of sodium methyl paraben preservative on the colour intensity of henna paste cones. The preservative has demonstrated very effective storage and colour retention for a long period of time. Yadav *et al.*[38] utilized the extract of spices which is commonly used for the preservation of poultry meat product and found very much effective in microbial safety of poultry meat product and improving the shelf life.

2.5 Application of Natural Dye as Colouring Material

Natural dyes can be applied in a very easy and low chemical consuming way. Dyeing of textile material with natural dyes can be carried out in a wide range of pH depending upon

the nature of the dyes. Their application on the textile material is a two step process which includes mordanting of textile material and application of dye on the fabric in general.

2.5.1 Different Mordants and Mordanting Methods

"The mordant word is derived from the Latin word 'Mordere' it means to adhere"[39]. Most of the natural pigments are not able to directly attach with the fabric material. They require some chemical or natural fixtures or metal salts, which can create adherence or affinity between natural dye particles and fabric surface. These cross linkers are called mordants[40]. A mordent or dye fixer is a substance that is used to form a coordinate complex with a dye to bind the colors on the fabric surface. Generally, the metal salts make the textile material more open, rough due to their corrosive action and makes them more favorable for the dye particles[41]. The natural ingredients containing tannins also works as natural mordants[42]. They make a connection between textile material and natural color and allow natural die particles to be easily fixed on textile material. Even they help dye particles which do not have affinity for textile material to get attached with the fabric[43]. In case of natural colors, if the textile material is able to color directly, then mordants help to produce deeper shades and darker colors[44]. Many scientists have reported their work on mordant application in natural dyeing process. Samanta *et al.*[45] studied the effect of single and double premordanting system and dyeing process variables on jute fabric dyeing with dye extracted from Palash flower petals commonly known as tesu. Srivastava *et al.*[46] studied the dyeing of silk fabric with the dye extracted from Himalayan Rhubarb using mixture of water and organic solvent methanol and reported maximum absorption. By using different mordants like Harda, Amla, Anar, Catechu, they obtained a wide range of colour shades. Singh *et al.*[47] evaluated the effect on the fastness properties of the dye extracted from evergreen shrub *Symplocos racemosa* (Lodh) found in Himalaya region. They used to dye wool fibres at optimized dyeing condition using various combinations of natural mordants (lemon juice) and synthetic mordants (i.e. $CuSO_4$, $FeSO_4$, $K_2Cr_2O_7$ and $SnCl_2$) in various ratios (3:1, 1:2 and 1:3). Different combinations give different colour shades and good to excellent fastness properties.

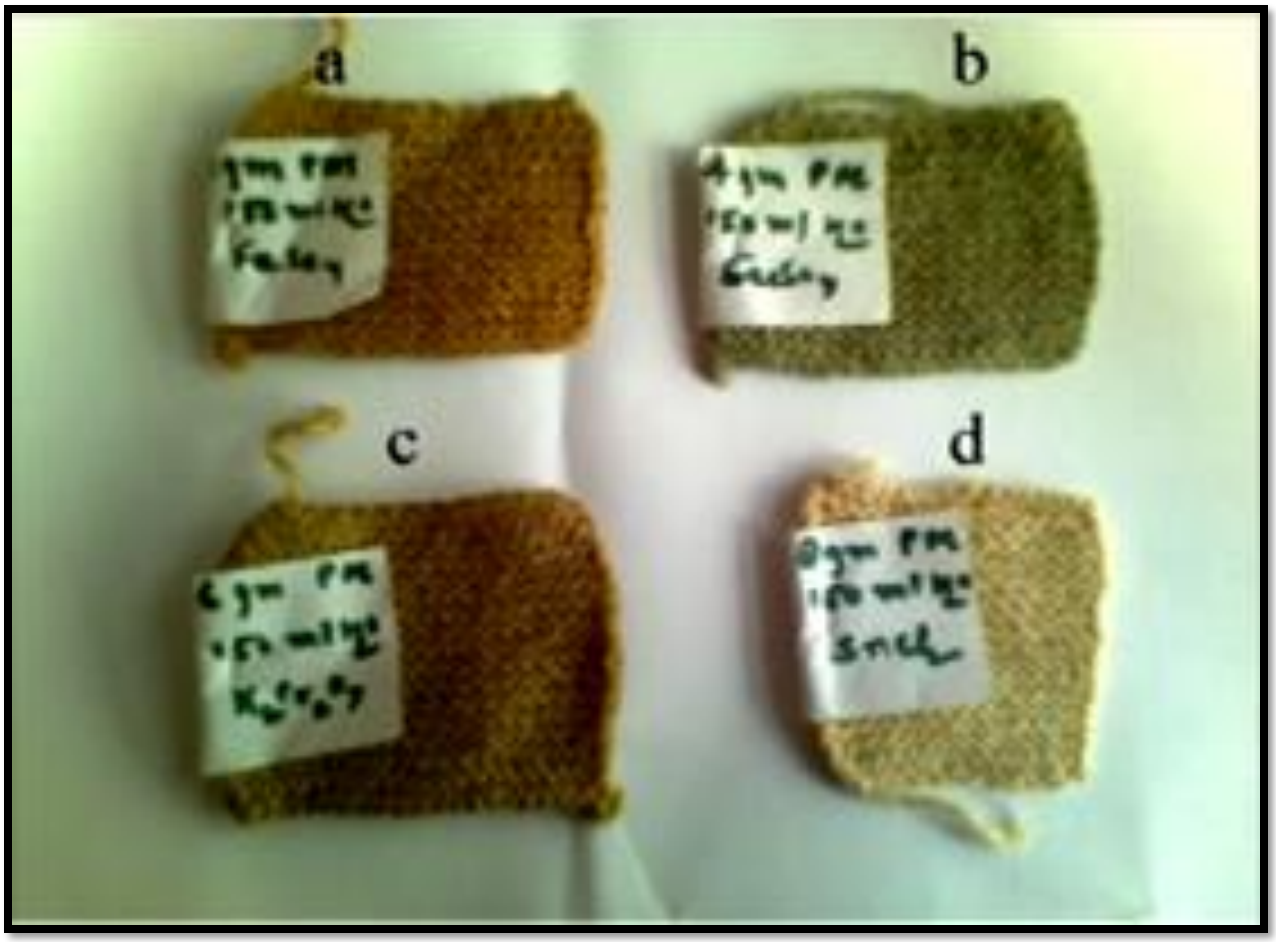

Figure 4. Dyed samples with and without mordanting using combination of synthetic and natural mordants (lemon juice)

Mohammad *et al.*[48] studied the application of natural dye extracted from madder root and heena through aqueous extraction in alkaline medium on woolen yarn using tin chloride II as a mordant and see its effect on colour strength, fastness properties, dyeability and antifungal activities and found good colour strength, fastness rating and improved antifungal activities. Onal *et al.*[49] observed the effect on dyeing of wool fabric, pretreated with Fermented dough extract FDE (a natural mordant) in weak acidic and neutral media. The dye was extracted from onion shell (*Allium cepa L.).* They have used different metal salt as a mordant and evaluated their effect on colour strength and fastness value. It has been observed that pretreated wool shows better colour strength and fastness value.

The mordants can be applied to textile materials in three different ways viz. pre mordanting method, simultaneous mordanting method and post mordanting method.

In **pre mordanting method** the mordant is applied to the textile material before dying. In this the fabric is first treated with the mordant in the separate bath and then the dyeing of the treated fabric is done in another bath. It is a two step process[50].

In **simultaneous mordanting method** the mordant is applied to the textile materials during dyeing process in the same dye bath[50].

In **post mordanting method** the mordants is applied to the textile material after dying. In the post mordanting method the fabric is first dyed and then treated the dyes fabric with the mordant to fix the colour or dye on it. It is also a two step process[50].

Lots of literature is available on the application of three different methods of mordanting. Zubairu *et al.*[51] studied the effect of various selected synthetic and natural mordant on cotton fabric dyed with the onion skin (*Allium cepa*). $FeSO_4$, $K_2Cr_2O_7$, $KAl(SO_4)_2$ and $CuSO_4$ were used as synthetic mordant and lemon and Aelo Vera were used as natural mordants. The cotton fabric was dyed by all three aforesaid mordanting techniques. In this work the simultaneous mordanting technique was found superior than the other two methods with respect to colour fastness properties of the dyed fabric. Swamy *et al.*[52] studied the dyeing of silk fabric using different mordants and different mordanting methods (pre-mordanting and Post-mordanting) for the dye extracted from Madhuca longifolia (Mahua leaves). They evaluate the fastness properties of dyed samples. The post mordanting method shows good washing fastness (rating 4-5) in this study. Janani *et al.*[53] studied the effects of different mordanting methods on cotton fabrics coloration with dye from Vernonia Amygdalina. It has been observed that post mordanting method gives better colour strength, brilliant colours and good fastness rating (i.e. 4-5). Raza *et al.*[54] utilize the onion skin to extract the dye and used for nylon fabric dyeing with different mordants like alum, potassium dichromate and copper sulphate through high temperature high pressure dyeing method. Afterward they analyse its effect on colour fastness properties and found good.

2.5.2 Effect of Dye Bath Condition on Natural Dye Application on Fabric

Dye bath condition includes dyeing time, dyeing temperature, pH of dye bath and material to liquor ratio of dye bath. Magnitude of dye penetration increases initially with time. However this initial increase follows a saturation plateau. Mostly used dyeing temperature range is from 60^{o}C to 90^{o}C. Sometimes dyeing is done even in boiling temperature of the used medium. Effect of heat upon dye uptake is very prominent. It shows improvement till the point of saturation reaches. Effect of pH is depends upon the natural dye source, material that is to be dyed and dyeing condition[55-56]. Lots of literature is available which have investigated the effect of dyeing conditions on dyeing of textile materials. Kashyap *et al.*[57] studied the cotton fabric dyed with coconut husk based natural dye. They extracted the dye from the coconut husk and applied to the cotton fabric under various dyeing conditions. They used two different mordant alum and vinegar and applied through different mordanting methods. Further they evaluated the dyed fabric against various fastness rating. They found satisfactory results. Giasuddin *et al.*[58] studied the extraction of dye from mango leaves to optimize the dye extraction conditions. Extracted dye is applied on silk fabric with and without mordant. They found significant colour strength and good washing and light fastness ratings (i.e. 4-5).

2.5.3 Application of Natural Dye on Different Textile Material

Dyeing of textile material with natural dyes can be carried out at acidic pH, alkaline pH or neutral pH depending upon the nature of the natural dyes and the fabric that is to be dyed[59]. Studies about dyeing of textile material with natural dyes are available in various literatures. Chavan *et al.*[60] studied the application of natural dye extracted from African Marigold on silk and cotton fabric. They dyed the cotton and silk fabrics under acidic and alkaline condition using alum and ferrous sulphate as a mordant and tested the dyed samples for various fastness rating. Grover *et al.*[61] extracted the natural dye from the Fire-Flame Bush floral part and studied their application on textile materials such as cotton, wool and silk. Saravanan *et al.*[62] studied the natural dye extracted from votiyar tree and applied on cotton fabric. The

cotton fabric is died under three different mordanting methods and tested for washing, light and rubbing fastness properties and found good. They have also studied the anti fungal and anti bacterial properties of the same. They have found them effective against, *Enterobactor* sp., *Staphylococcus Aureus,* and *Staphylococcus* sp. bacterial pathogens and also against *Candida* sp., *A. Flavus and A. Niger* fungal pathogens. Jothi *et al.*[63] uses African marigold to extract the dye to evaluate its dye potential and applied on cotton and silk fabric and studied its dyeability and colour fastness properties.

2.5.4 Application of Natural Dyes in Different Combinations

The Natural dye can be applied to the textile material in various combinations. Various dyes can be taken in definite proportion and mix together to dye textile materials to get various colour strength and fastness ratings. It is also helpful to get suitable combination of natural dyes which can be mixed to get the best results in terms of colour strength and various fastness ratings. Some scientists have worked on this area. Samanta *et al.*[64] used the mixture of Jackfruit Wood with the various other natural dyes extracted from Marigold, Red Sandal Wood, Manjistha, Sappan wood and Babool for dyeing the jute fabric and studied the colour performance and dye compatibility of these dye mixture on jute fabric. This study helps in finding the compatible and most suitable binary mixture of jackfruit wood with Marigold, Red Sandal Wood, Manjistha, Sappan wood and Babool for dyeing jute.

2.5.5 Unconventional Application of Natural Dyes

Growing demands of consumers for the ecofriendly textile and variety of shades led to the development of more efficient and more energy efficient process that can be able to develop variety of shades in the field of natural dyes[65-67]. Other than this conventional colourful use these sometimes applied for some of their additional properties. Camphor plants extract posses antibacterial properties as well as UV resistance along with traditional colouring properties.

Jiang *et al.*[68] had utilised this extract to impart antibacterial and UV resistance effect on the silk fabric. They had also analyzed the colour strength in terms of fastness rating and found excellent (4-5). Further they studied the ultraviolet protection properties from UVA and UVB radiation, antibacterial properties against the bacterial Staphylococcus aureus, Candida albican and Escherichia along with colour strength values (K/S Value) and colour fastness properties of the silk fabric. Mirjalili *et al.*[69] studied the extraction and characterization of natural dye from green walnut shells and its use in polyamide dyeing. However simultaneously they have checked its antibacterial properties.

In conclusion it can be summarised that various natural resources had been utilized as dye source. But the stock of the nature is limitless. Hence, new sources with easy availability, low cost and good shelf life is always in demand for textile dyeing. In this context beetroot is one of the strong candidates. Brief biological information about beetroot is table 3 given below.

Table 3: Biological information about beetroot[70].

Name	Beetroot
Scientific Name	Beta vulgaris
Plant characteristics	It is a plant with leafy stem 1-2 m tall. The leaf is heart shaped which is 5-20 cm long. It has a very small flower around 3-5 mm diameter with five petals having dense spikes and green or tinged reddish colour.
Nutrient Value	The main ingredients in beetroot are carbohydrates (50%), Dietary Fiber (11%) amino acids (10%), betaine (4%), proteins (10%), vitamins A, B and C and minerals (15%).
Medicinal properties	Digestive, laxative, anti parasitic, antifungal and antibacterial
Traditional use	Food colouring agent, food ingredient

Beetroot

Though beetroot is being utilized as food colouring agent and food ingredient. But it hasn't been explored much as textile colouring agent. Therefore beetroot has identified as the key ingredient for the proposed research.

3. Rationale of Research.

At the end of the 20th century, many countries of Europe and Asia including India have banned certain synthetic pigments due to their harmful impact on nature and their habitants. In the 21st century, an increasing awareness and consciousness towards environment have pushed scientists and researchers to put the concentration on natural dye sources. Recently, the use of natural dye is increasing due to various stringent environmental standards imposed by various concerned authorities. The availability of scientific literature regarding natural dye and their application on textile material is much abundant. Those reported research has pointed out here some limitation of natural dye. One of the biggest challenges for natural dyestuff application is the limited sources for red and blue colors[71]. Natural colors derived from beetroot can be used as red dye source as they are cheap, abundant and easily available all over India. Their renewable source makes it environment-friendly, biodegradable, non allergic and do not cause any human and environmental damage. Literature on beetroot as natural dye source for textile material is rather scanty and infrequent. Red beetroot (*Beta vulgaris)* belongs to Chenopodiaceous family[72]. The literature shows that this red pigment

extracted from beetroot is vastly used as antimicrobial and antioxidant agents in food, medicine and allied industries and as a dyeing source in leather industries[77]. The main pigment found in beetroot is betalain, which is one of the richest sources of betanin pigment. This betanin pigment is mainly responsible for red color[73]. Beetroot colour is traditionally used for centuries as a natural colouring agent in meat product[74]. Betalain has been successfully used in commercial food color operations for many years and it is an important source of coloring agent in the food industry[75]. There has been a lot of work on potential use of beetroot colour as a food and cosmetic natural colouring material[76]. Application of beetroot dye on the leather is also suitable[77]. The beetroot extract as a colorant is used in cream cheese. Also it has a great potential as alternative colouring for food[78]. Even betanin can be used as a colouring and flavoring agent for sauces, tomato paste jams and jellies ice creams and sweets[79].Very limited research on extraction of dye from beetroot and its application on textile materials have been reported till date. Summoogum-Utchanah *at el.*[80] reported the work on extraction of natural dye from beetroot and turmeric. Kumar *at el.*[81] studied the extraction and analysis of natural dye from ten plant source, one of which was beetroot and applied on cotton and jute yarn. Tasneem *at el.*[82] studied the aqueous extraction of beetroot natural dye from the beetroot and its application by using different mordants on wool and cotton threads. So the present study is mainly focused on the optimization of the extraction of beetroot dye and its potential application on protein fibre using different natural and synthetic mordants with respect to extraction condition and extraction medium.

4. Objective of the Study

From the above mentioned literature review the objective of the present study has been set to Optimize the extraction process of beetroot dye and its potential application on protein fibre using different natural and synthetic mordants with respect to extraction conditions and extraction medium.

So to attain this objective following goals are framed.

1. Optimization of extraction for maximum recovery of Beetroot natural dye at different extraction conditions.
2. Study of extracted Beetroot natural dye shelf life with or without natural preservative.
3. Optimization of dyeing wool/Silk fabric with extracted natural dyes with respect to maximum colour strength.
4. Study of colourfastness of dyed wool/silk fabric against washing, and rubbing action.

5. Material and Methods

1.1 Materials

1.1.1 Raw Materials

- **Dye source**: Raw beetroot
- **Fabric :** 100% wool/silk fabric

1.1.2 Auxiliary Chemicals

1.1.2.1 Dye Extraction

- Acetic acid (CH_3COOH) or citric acid ($C_6H_8O_7$) will be utilised for acidic pH adjustment, sodium hydroxide (NaOH) or sodium carbonate (Na_2CO_3) will be utilised for basic pH adjustment.
- pH capsules for calibration of pH Meter.
- Water – Distill water or deionised water

1.1.2.2 Scouring

- Sodium carbonate (Na_2CO_3): sodium carbonate will be used as a scouring agent.
- Non ionic detergent: It will be used as a wetting agent.

1.1.2.3 Bleaching

- Hydrogen Peroxide (H_2O_2): It will be used as oxidative bleaching agent.
- Thiourea (CH_4N_2S): It will be used as reductive bleaching agent.

1.1.2.4 Mordanting

- Mordants:
  - Alum [$KAl(SO_4)_2$] will be used as inorganic mordant.
  - Lemon ($C_6H_8O_7$) or hara will be used as organic mordant.
  - Ferrous sulphate ($FeSO_4$) or potassium dichromate ($K_2Cr_2O_7$) will be used as synthetic mordant.
- Water: RO water.

1.1.2.5 Dyeing

- Natural Dyes: Beetroot (*Beta Vulgaris*) etc.
- pH adjusting ingredients will be same as used in dye extraction.
- For dye exhaustion: sodium chloride (NaCl).
- Water: RO water.

1.1.2.6 Washing

- Water: RO water.

1.1.3 Glass ware and auxiliary material

- Beaker (75 ml – 1000 ml)
- Glass rod (short and long)
- Glass bottle with cap (30 ml – 500 ml)
- Pipette with pipette pump (1 ml – 10 ml)
- Measuring cylinder (100 ml)
- Volumetric flask (100 ml)

1.1.4 Instruments

- Dye bath
  - Maker's name: Paramount creative ventures Pvt. ltd.
  - Temperature range: Room temperature to 100^{o}C)
  - Application: Dye bath can be utilised for scouring, bleaching, mordanting, dyeing and washing.

- Dye extraction setup: This will be assembled as per requirement. The setup will comprise of heating mantle, flat bottom beaker with lid, magnetic stirrer and thermometer. All these components are dedicated for homogeneous temperature distribution throughout the liquid for even dye extraction at boiling point of the respective medium.
- Spectrophotometer
    - Maker's name: SHIMADZU FTIR
    - Model no: 8400 F,
    - Frequency range visible spectra from 400 nm to 700 nm.
    - It will be used for colour strength measurement in liquid medium and on the fabric.
- pH meter
    - Maker's name: Systronics
    - Model no: MK VI
    - It will be used for adjusting the pH of extraction bath, pretreatment processes bath and dye bath.
- Crock Meter
    - Maker's name: Paramount creative ventures Pvt. ltd.
    - Model: Analog
    - It will be used for the assessment of rubbing fastness of dyed samples.

1.2 Methods

1.2.1 Dye extraction

- Natural dye source will be preprocessed to make it clean by washing and devoid of any unknown particles of the source itself or the plant. Cleaned natural dye source will be processed into small pieces for better dye extraction. The chopped natural dye source will be put into the flat bottom beaker with deionised water or alcohol as extraction medium. Then the system will be brought to boiling condition for the dye extraction. Different extraction condition for dye extraction is as follows (Table 4)

Table 4: Dye extraction conditions[16-20].

S. No.	Component	Particular	Quantity
1	Dye source	Raw Beetroot	200 g
2	Extraction medium Volume	Deionised Water or Alcohol	1000 ml
3	Material to liquor ratio	----------	1:5
4	pH	----------	3-11
5	Temperature	----------	60-90^{o}C
6	Time	----------	60-120 min

1.2.2 Pretreatment of Fabric Sample:

- **Scouring: S**couring is done to remove wax. In this process the fabric sample is heated in the solution prepared from 0.5gpl Na_2CO_3 and 2gpl non ionic detergent for about 30 minutes at 45-50^{o}C[49].
- **Bleaching (If Applicable):** Bleaching is done to reduce pigment density of the fabric sample. The bleaching of fabric sample will be done with the help of H_2O_2 at alkaline pH (below 10) for 1 h at 60^{o}C. Thiourea is then added to the bath and heated it for 25 min at 60^{o}C after adjusting the bath pH to around 4.5-5.5. The fabric is then washed and rinsed thoroughly[83].

1.2.3 Dyeing Process

- **Mordanting Application:** pre mordanting method will be applied for the dyeing. In this process the fabric is put into the flat bottom beaker for pre mordanting using different mordants separately. The mordant used in the process will be alum (inorganic), lemon or harda (organic), ferrous sulphate or potassium dichromate (synthetic) and RO water is used as mordanting medium. The fabric is then mordanted by keeping the different mordanting condition as given in the table below (Table 5).

Table 5: Mordanting conditions[45-50].

S. No.	Component	Type	Quantity
1	Fabric	Wool /Silk	5 g
2	Dyeing medium Volume	RO Water	1000 ml
3	Material to liquor ratio	----------	1:20
4	Mordant	Alum	2-5% OWF (on the weight of the fabric)
		Lemon or Harda	
		Ferrous sulphate or potassium dichromate	
5	Temperature	----------	60-90^{o}C
6	Time	----------	60-120 min

- **Dyeing of the Fabric:** The fabric is kept into the beaker for dyeing. RO Water is used as dyeing medium. The fabric is dyed in Dyebath by keeping different dyeing condition given in the following table (Table 6).

Table No. 6: Dyeing conditions[55-56].

S.No.	Component	Type	Quantity
1	Fabric (Wool/Silk)	Premordanted with Alum	5 g
		Premordanted with Lemon or Harda	
		Premordanted with Ferrous sulphate or potassium dichromate	
2	Dyeing medium Volume	RO Water	1000 ml
3	Material to liquor ratio	----------	1:20
4	Mordant	Alum	2-5% OWF
		Lemon or Harda	
		Ferrous sulphate or potassium dichromate	
5	NaCl	----------	1-2 g/l

6	Temperature	----------	60-90^{o}C
7	Time	----------	30-90 min

- **Washing of Dyed Fabric Samples:** The fabric will be washed in RO water at 60-90^{o}C for 15-30 min with a Material to Liquor ratio of 1:20.

1.2.4 Characterization (spectrophotometric characterization)

- **Characterization and Standardization of Dye Extraction:**

  Characterization and standardization of extracted dye solution will be done with the help of visible range spectrophotometer. The spectophotometric principle of Beer Lambert law is used to calculate the concentration of the dye solution using the given formula[31]

  $$\text{Log } (I/I_0) = \epsilon dc \text{ ----------} \quad (1)$$

  Where,

  I = Intensity of the light source

  I_0 = Intensity of the light passed through the solution

  ϵ = Molar absorptivity or molar absorption coefficient.

  d = Length of solution the light passes through

  c = Concentration of the dye solution

  i. First step is to prepare a calibration plot against known weight of raw beetroot with the help of spectrophotometer in transmittance mode.

  ii. In the second step extraction of dye from raw beetroot by varying the parameter like temperature, time, pH and extraction media, are characterized and quantified with the help of above mentioned calibration plot and spectrophotometric data.

- **Characterization and Evaluation of Colour Strength of Dyed Fabric.**

  i. Direct method: In this method dyed fabric is directly analyse through spectrophotometer in reflectance mode with the help of above mentioned calibration plot. This method is preferred for rubbing fastness assessment.

ii. Indirect method: In this method colour strength of original Dyebath before (A) and after dyeing (B) along with washing bath (C) is analyse through spectrophotometer and amount of dye transferred to the fabric is calculated using the formula {A – (B + C)}. This method is preferred for dye transfer analysis and washing fastness assessment.

2. Plan of Work

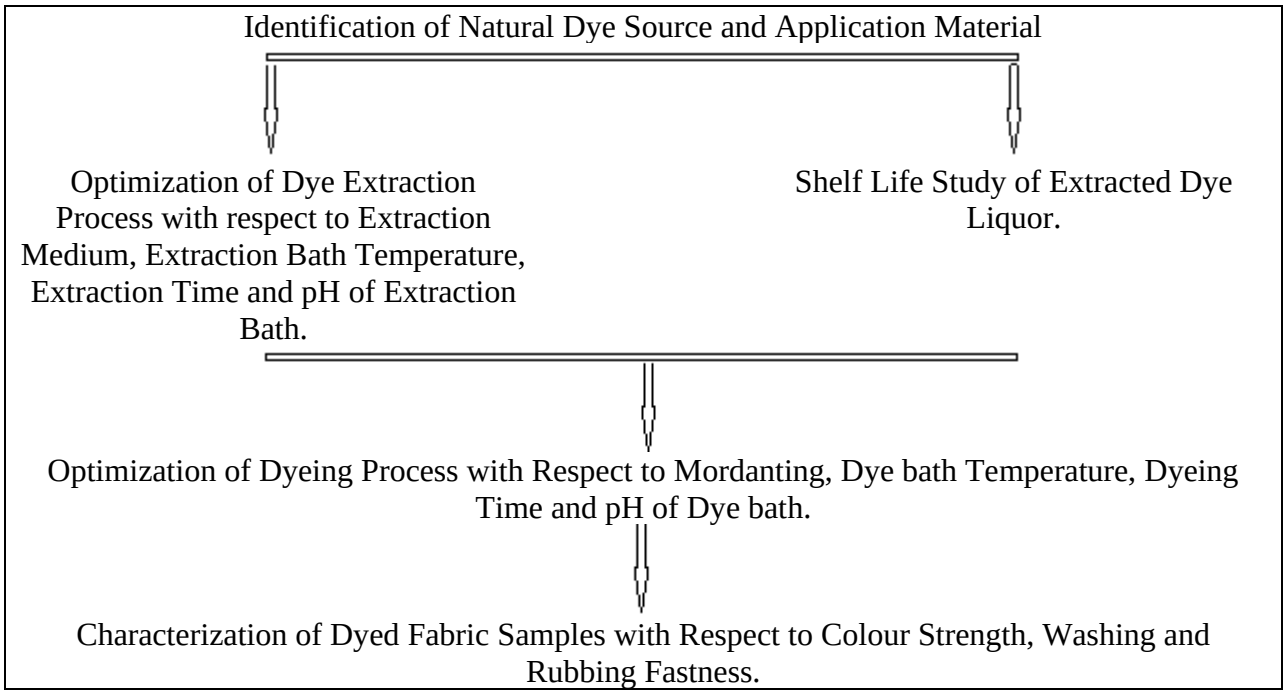
Identification of Natural Dye Source and Application Material
Optimization of Dye Extraction Process with respect to Extraction Medium, Extraction Bath Temperature, Extraction Time and pH of Extraction Bath.
Shelf Life Study of Extracted Dye Liquor.
Optimization of Dyeing Process with Respect to Mordanting, Dye bath Temperature, Dyeing Time and pH of Dye bath.
Characterization of Dyed Fabric Samples with Respect to Colour Strength, Washing and Rubbing Fastness.

Figure 5. Flow chart for plan of work.

7. Impact of the Proposed Research Work in the Field of Science and Technology as Well as on Society.

Environmental considerations have become an important factor all over the world. Even awareness towards environment among peoples and laws improvised by several governing agencies all over the world are forcing every manufacturing institute to shift to a greener technology. In this shift textile is also included. Today World is facing the acute environmental problem like environmental pollution, depletion of ozone layer green house

effect and last but not the least waste disposal problem[84]. Synthetic dyes are the chemicals which have adverse effect on environment as well as human being. Moreover dyeing is the most water consuming process so after dyeing process when this waste water containing chemical is disposed in to the environment it will lead to the serious environmental pollution and health problems. So promoting the use of natural dyes in textile dyeing and printing will definitely be an advantage as it will be helpful in reduction of environmental problems and maintaining ecological balance and ultimately gives benefit to the society. Our research is on beetroot dye which is totally ecofriendly and obtained from natural renewable resources. Moreover this study is aimed to provide the knowledge about the process of maximum extraction of beetroot natural dye by optimizing the pH of dye extraction bath, optimization of dying process of dyeing wool/silk fabric using natural and synthetic mordant to evaluate the dyed fabric for colour strength and various fastness ratings and improvement of shelf life of beetroot natural dye using natural preservatives. Also it will help the maximum extraction of dye from the natural dye source and their by increasing the profitability of the people involved in the natural dye business.

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